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Importance of annual monitoring for evaluating the direct nitrous oxide emission factor in temperate mono-rice paddy fields

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ABSTRACT

In temperate mono-rice paddy fields, rice is cultivated for 100–140 days under submerged conditions during the summer period, and thereafter, the field remains under dry conditions during the winter and spring seasons. However, the early developed nitrous oxide (N_2O) emission factor (EF) was only based on seasonal (rice cropping period) N_2O fluxes, which resulted in lower N_2O EF than the default value (0.3%) used by the Intergovernmental Panel on Climate Change (IPCC). Furthermore, the long fallow season may be favorable for nitrification and substantially result in increased N_2O emissions. A two-year field experiment was conducted to evaluate the effect of N_2O emissions during the dry fallow season on the annual N_2O EF. The N_2O emission rates were sequentially characterized during the rice cropping and the fallow season under four different levels of nitrogen (N) fertilizer for rice cultivation. The urea was applied at four different (0, 45, 90 and 180 kg N ha^{-1}) levels, and rice was cultivated under submerged conditions during late May to early October. The seasonal N_2O fluxes during the rice cropping and fallow seasons clearly increased with increasing N application rates. In the N fertilized plots, the mean N_2O emission rates were higher during the fertilized cropping season than the fallow season, but the seasonal fluxes were much higher during the unfertilized fallow season, due to the long dry period. The seasonal N_2O EF, which was estimated by the increased N_2O flux with N fertilizer, was only 0.0015–0.0017 kg N_2O-N kg^{-1} N during rice cropping. However, the annual N_2O EF combining the two seasonal N_2O fluxes markedly increased to 0.0028–0.0031 kg N_2O-N kg^{-1} N, which was very close to the N_2O EF of the IPCC. Conclusively, the N_2O EF in mono-rice paddy fields should be developed using the annual N_2O fluxes and not only the cropping seasonal N_2O fluxes.

1. Introduction

Over the past 50 years, global temperature has increased at the fastest rate in recorded history, mainly due to increasing greenhouse gas (GHG) emissions (Chen et al., 2010). Carbon dioxide (CO_2), methane (CH_4) and nitrous oxide (N_2O) are the major GHGs (Myhre et al., 2013) that cover approximately 63%, 24% and 3% of total GHG emissions, respectively (Forester et al., 2007; Luo et al., 2013; Jiang et al., 2016). Among these GHGs, N_2O has the greatest radiative force on a per mass basis with a global warming potential (GWP) of 310 times that of CO_2 over a 100-year period (IPCC, 2007; Jahangir et al., 2013). Additionally, N_2O has become the most important substance contributing to ozone layer depletion after chlorofluorocarbons were phased out (Ravishankara et al., 2009).

In agricultural soils, N_2O emissions are mainly produced by the extensive chemical N fertilizer use and increasing manure inputs (IPCC, 2007). Globally, agricultural fields account for almost 60% of the total anthropogenic N_2O emission (Bouwman et al., 2002; Smith et al., 2007). To meet the increasing global demand for food, N_2O emissions are predicted to increase by approximately 35–60% by the end of 2030 (Smith et al., 2007). To form a reasonable strategy to reduce N_2O emission from the agricultural sector, the precise estimation of N_2O fluxes should be first established. However, N_2O estimation has still been uncertain, due to insufficient field measurements for temporal and

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spatial representations of soil, agricultural management practices and climate (IPCC, 2001). Nitrous oxide emission from agricultural fields differs by several-fold or even orders of magnitude across different experimental sites and seasons (Dobbie and Smith, 2003; Wagner-Riddle et al., 2008; Wang et al., 2011). Therefore multiyear, continual measurements in situ are crucially important for eliminating the uncertainty of N_2O emissions at the regional and global scales.

In temperate mono-rice paddies, rice is generally cultivated under flooded conditions for 100–140 days while the soil is preserved under dry fallow conditions after rice harvesting for > 220 days. Most of the studies related to N_2O emissions from various rice cultivation systems focused on the irrigated rice cropping season (Cai et al., 1997; Khalil et al., 2008; Xu et al., 1997). In the current mono-rice paddy field, high N_2O emissions may occur during the dry fallow season due to a long period with a high nitrification rate under aerobic soil conditions. However, our understanding of N_2O emissions during the dry winter season is poor due to the scarcity of available field N_2O measurements (Zhou et al., 2014). Several studies clearly showed higher N_2O fluxes in the fallow season than in the irrigated rice cropping period (Zhou et al., 2014). These results also emphasize that field measurement of N_2O emissions from mono-rice cropping systems must be conducted based on the whole year, not only the irrigated rice cultivation period.

To develop the research protocol to determine the N_2O EF in temperate mono-rice paddy soil (in rice cropping and fallow seasons), four different levels of N fertilizer were applied for rice cultivation and the N_2O emission rates were consecutively monitored for two years. Finally, the N_2O EF was compared between the cropping and fallow seasonal and annual bases.

2. Materials and methods

2.1. Experimental plot preparation and rice cultivation

The study was conducted in the agronomic rice field of Gyeongsang National University, Sacheon, the Republic of Korea (35° 8′56″N, 128° 5′46″E), during two years (2014 and 2015). The selected site was a typical rice paddy, and only rice has been cultivated here for more than fifty years. The selected soil belongs to the *Pyeongtaeg* series (fine-silty, mixed, nonacid, mesic Typic Haplaquent). Before the experiment, the physiochemical properties of soil were pH 5.6 ± 0.2 (1:5 with H_2O), total C 8.9 ± 0.6 g kg^{-1} and total N 0.65 ± 0.08 g kg^{-1}.

The study plots (each plot: 10 m × 10 m size) were installed in a randomized block design with three replicates per treatment. To avoid nutrient-mixing effects, the experimental plots were isolated from each other by inserting concrete barriers (soil depth of 30 cm) as buffer zones (60 cm). Nitrogen fertilizers were selected as the main treatment with four different levels (0, 45, 90, and 180 kg N ha^{-1} as urea). Potassium (58 kg K_2O ha^{-1} as potassium chloride) and phosphate (45 kg P_2O_5 ha^{-1} as superphosphate) were applied at the same level in all treatments. Fertilizer application was split into three times according to the Korean standard method (RDA, 1999). The basal fertilizers (50%, 100% and 70% of N, P_2O_5 and K_2O application doses, respectively) were applied one day before rice transplanting. Nitrogen was split into 20% and 30% of the application dose at the 14th and 42nd day after transplanting, respectively, and the remaining potassium (30%) was additionally applied at the 42nd day after transplanting.

Twenty-one day-old rice seedlings (*Sin-Dongjin cultivar*, *Japonica*) were manually transplanted (30 cm × 15 cm) in May, and rice was harvested in the first week of October. The irrigated water level was maintained at a depth of 5–8 cm over the surface during the rice cultivation season and was drained one month before harvesting. After rice harvesting, the field was maintained without any management during the cold and dry fallow season, from late October to early the next May.

2.2. Monitoring the physical and chemical properties of the soil

Soil temperature was recorded throughout the rice cropping and fallow seasons by a thermometer placed in the soil at a depth of 10–15 cm. Soil redox potentials (Eh values) were measured only during the waterlogged rice cropping season by an Eh meter (PRN-41, DKK-TOA Corporation) at the aforementioned depth. In addition, soil moisture contents were monitored only during the dried fallow season using data logger sensors (EM50 Data logger, Decagon Devices, USA).

To monitor the changes in the inorganic N (NH_4^+-N and NO_3^--N) concentration in soils, moist field soils were collected at the surface layer (0–15 cm) five times during the cropping season and eight times during the fallow season for two years. Fresh soil samples (10 g) were extracted using 50 ml of 2 M KCl solution, and the NH_4^+-N and NO_3^--N concentrations in the extracts were quantified by the brucine method (Jenkins and Medsken, 1964) and the indophenol blue method (Kempers and Kok, 1989), respectively.

2.3. Monitoring of N_2O emission rates, seasonal N_2O fluxes and the emission factor (EF)

The N_2O emission rates were monitored using a static closed chamber method (Rolston, 1986; Cuello et al., 2015) during the two years. A cylindrical acrylic chamber (H. 20 cm, D. 24 cm) was installed permanently in the ground at the base chamber between each rice plant (Kim et al., 2014; Kim et al., 2017). The bottom chamber has two holes in the bottom for water movement during the rice cultivation period, which were blocked with rubber stoppers after being drained. The chambers were kept open except for gas sampling.

Gas sampling was conducted three times per day (8:00 am, 12:00 pm, and 6:00 pm) to get the daily mean N_2O emission rates with a one-week interval. The gas was collected at an interval of 30 min (0 and 30 min) after covering the base chamber with the opaque top chamber (H. 20 cm, D. 24 cm) using 50-ml air tight syringes; collected samples were immediately transferred into a vacuum vial (20 ml).

The collected gas samples were analyzed for the N_2O concentration with gas chromatography (GC-2010, Shimadzu, Japan) equipped with an electron capture detector (ECD) and Porapak Q column. The temperature of the column, injector and detector were adjusted to 35 °C, 200 °C and 300 °C, respectively. Helium was used as the carrier gas. A carrier gas filter, installed in the gas supply line, is capable of trapping oxygen, moisture and organic compounds.

The N_2O emission rates were calculated by comparing the increase in each gas concentration per chamber surface area during the gas collecting time interval (Shen et al., 2014) (Eq. (1)):

$$R = \rho \times (V/A) \times \Delta C \times (273/T) \tag{1}$$

where R is the N_2O flux (μg m^{-2} h^{-1}), ρ is the gas density N_2O under a standardized state (g m^{-3}), V is the chamber volume (m^3), A is the chamber surface area (m^2), ΔC is the increase rate of the N_2O concentration in the chamber (μg m^{-3} h^{-1}), and T (absolute temperature) is 273 + the mean temperature (°C) of the inner chamber during the sampling period.

The seasonal N_2O flux was determined as follows (Eq. (2)) (Singh et al., 1999):

$$\text{Seasonal } N_2O \text{ flux} = \Sigma_i^{\,n} (R_i \times D_i) \tag{2}$$

where R_i is the N_2O emission rate (μg m^{-2} h^{-1}) in the ith sampling interval, D_i is the number of days in the ith sampling interval, and n is the number of the sample size.

Based on the methodology of the IPCC, which defines the N_2O EF as the linear relationship between N_2O emissions and N fertilizer (IPCC, 2006), the EFs were calculated from the linear regression slopes obtained between the total N_2O fluxes and various N application levels (Kim et al., 2017). These slopes were considered as the N_2O EFs from N fertilizer applied soils.

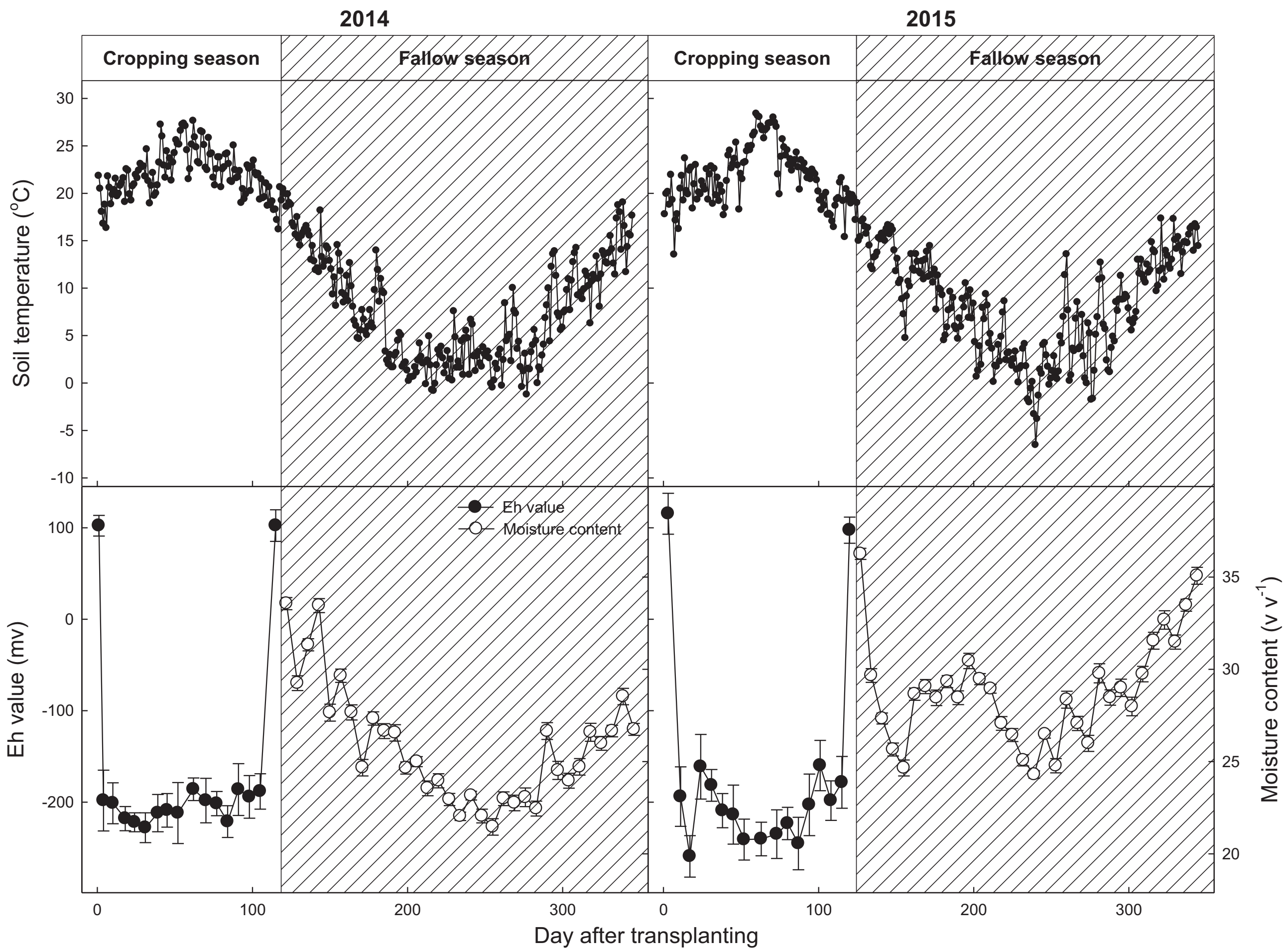

Fig. 1. Variation of soil temperature, Eh values and moisture content in rice paddy soil during the cropping and fallow seasons (note: Eh values were measured only during the flooded rice cropping season, and moisture content was measured only during the dry fallow season).

2.4. Statistical analysis

Statistical analysis for the experimental data was computed using the SPSS software package (IBM SPSS Statistics 23). Differences in the means were determined using Tukey's honest significant difference (HSD) test using one-way analysis of variance (ANOVA) if the F-test was significant at the $p < 0.05$ probability level. Linear regression and correlation analyses were performed to evaluate the relationships between response variables.

3. Results

3.1. Soil temperature and moisture regimes

Soil temperature change patterns were not significantly different between the 1st and 2nd years and did not differ among the treatments (Fig. 1). Temperatures sharply increased after rice transplanting, peaked at the flowering season (the end of July) and slowly decreased thereafter. The lowest temperature was recorded in mid-January in the winter season and, thereafter, slightly increased again. The mean soil temperature was 21.8–22.0 °C during the rice cropping season, a marked difference from 7.6 to 8.4 °C during the fallow season.

The Eh values similarly changed during the rice cultivation periods in both years, but not among the different treatments (Fig. 1). These sharply decreased after flooding during rice cultivation and then stabilized under an extremely reduced condition (near minus 200 mV) throughout the rice cropping periods until drainage for harvesting.

The soil moisture content similarly changed during the dry fallow season for two years, but not among the different treatments (Fig. 1). It gradually decreased after harvesting but clearly increased with precipitation from early April to transplanting season.

3.2. Nitrous oxide emission rates

While nitrous oxide emission patterns similarly changed in both years, its emission rates largely differed between the waterlogged rice cropping and the dry fallow season (Fig. 2). During rice cultivation, the N_2O emission rate significantly increased with increasing N fertilizer application. Nitrous oxide emission rates highly increased right after the basal (one day before transplanting) and split N fertilizer applications (14th and 42nd days after transplanting). Thereafter, N_2O emission rates gradually decreased until the harvesting stage. In comparison, an insignificant difference in N_2O emission rates was observed among the treatments during the dry and cold fallow seasons. These clearly decreased with declining temperature until mid-winter and thereafter gradually increased as the temperature grew warmer.

Mean N_2O emission rates were higher during the warm and fertilized rice cultivation period than during the cold and unfertilized fallow season under the same level of N fertilizer. For example, in the no N fertilizer treatment (hereafter, control treatment), the mean N_2O emission rate was 0.167–0.173 mg N_2O-N m^{-2} day^{-1} during the rice cropping season. This was slightly higher than 0.154–0.160 mg N_2O-N m^{-2} day^{-1} during the cold fallow season but not statistically significant. The difference in mean N_2O emission rates between rice cultivation and fallow seasons became much higher with increasing N application level. For example, at the recommended N fertilizer

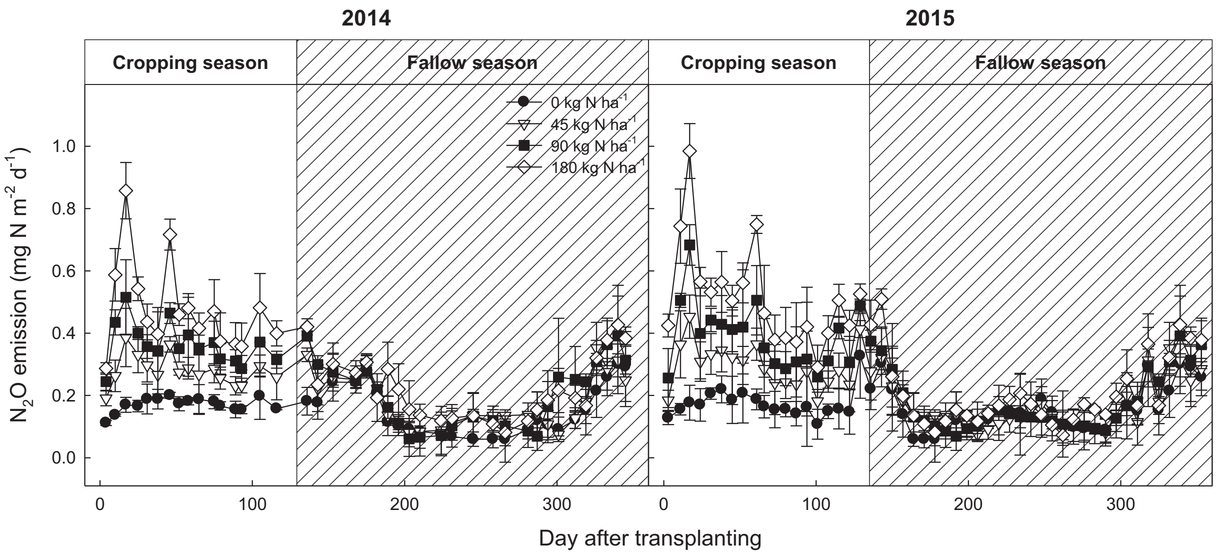

Fig. 2. Changes in the N_2O emission rate from the mono-rice field under different N application rates in the cropping and fallow seasons during the 2-year study.

treatment (90 kg N ha^{-1}), the mean N_2O emission rate was 0.284–0.328 mg N_2O-N m^{-2} day^{-1} during the rice cropping period, which was much > 0.225–0.241 mg N_2O-N m^{-2} day^{-1} in the dry fallow season.

Irrespective of the study year, fallow season N_2O emission rates showed higher positive and significant correlations with soil temperature ($p < 0.001$) and soil moisture content ($p < 0.001$) than cropping season emission rates. Similarly, N_2O emission rates during the cropping season were also significantly and positively correlated with temperature ($p < 0.01$) and slightly negatively correlated with the Eh value ($p < 0.01$) (Table 2). Inorganic N concentrations (NO_3^--N and NH_4^+-N) were very strongly correlated with N_2O emission rates; NO_3^--N ($p < 0.001$) showed a higher correlation than NH_4^+-N ($p < 0.01$) regardless of the seasons and years (Table 2).

3.3. Soil inorganic N contents

Two different types of inorganic N contents (NH_4^+-N and NO_3^--N) were changed similarly during the two years of the field study. However, both the NO_3^--N and NH_4^+-N concentrations showed large dissimilarities between the rice cultivation period and fallow season (Fig. 3). During cropping seasons, the NH_4^+-N and NO_3^--N contents were highly influenced by N fertilizer levels and its application timing. The inorganic N content significantly increased with increasing N application levels and became greater right after application at the 14th and 42nd days after rice transplanting (Fig. 3). Thereafter, the inorganic N content clearly decreased with time. In comparison, very low levels of inorganic N content were detected at the fallow season. However, slightly higher NO_3^--N and NH_4^+-N contents were observed in higher N fertilizer applied soil. In particular, most of the inorganic N was represented by NO_3^--N type in the dry fallow season.

3.4. Nitrous oxide emission factor

Nitrous oxide EFs for the flooded rice cropping, dry fallow season and entire year were calculated using the linear relationship between total N_2O fluxes and N fertilizer levels (Fig. 4). The seasonal N_2O EFs ranged from 0.15–0.17% and 0.13–0.14% in both years under the rice cropping and fallow seasons, respectively (Fig. 4). These EF values were only 46.3–56.7% and 46.7–50.0% of the IPCC default value during the rice cropping and fallow seasons, respectively (Table 2). The overall EFs for the entire season that account for the cumulative N_2O fluxes were 0.28–0.31%, close to the IPCC default value (0.3%) for rice paddy (Table 2).

4. Discussion

Nitrification and denitrification are the main processes that produce N_2O in the soil (Dobbie and Smith, 2003; Henault et al., 2012); nitrification was the main process responsible for N_2O production, which accounted for > 70% of the total N_2O flux (Stevens et al., 1997). In this study, N_2O emission rates were generally low during the flooded rice cropping period because nitrification was either retarded or inhibited due to the lack of oxygen concentration during the flooded period (Akiyama et al., 2005; Zou et al., 2007). For example, the mean N_2O flux in the control plot (0 kg N ha^{-1}) during the cropping season was 0.17 mg N_2O-N m^{-2} d^{-1} in the rice paddy (Fig. 2), not comparable with 0.66 mg N_2O-N m^{-2} d^{-1} in the maize upland soil in the same region (Kim et al., 2017). The relatively low N_2O fluxes during the flooded rice cropping season could be attributed to the low soil redox potential (Eh), which is unfavorable for nitrification. The peaks for N_2O flux after N fertilizer application during the rice cropping season (Fig. 2) are in agreement with that of Bronson et al. (1997) and Zou et al. (2005) who stated that the applied N undergoes nitrification and denitrification processes that finally lead to the production of N_2O.

Soil temperature and moisture and inorganic N content are the main factors that increase N_2O emission from arable land (Liu et al., 2014; Das and Adhya, 2014; Pajares and Bohannan, 2016). In this study, soil temperature, moisture and inorganic N content were also highly correlated with N_2O emission during both seasons (Table 2). In particular, the soil temperature and inorganic N content were highly and positively correlated with N_2O flux both during the cropping and fallow seasons. The Eh value showed a negative correlation with N_2O flux during the cropping season due to the reduced oxygen concentration in the soil (Akiyama et al., 2005; Zou et al., 2007), but a positive correlation between moisture content and N_2O flux was observed during the fallow season.

Nitrous oxide emission is largely dependent on N input as well as water management in croplands (Zou et al., 2009; Liu et al., 2010). In addition to N input during the rice cropping season, large amounts of N_2O emissions might be expected during the long, dry fallow season; the seasonal effect on N_2O emission was not considered previously for estimating the direct N_2O EF in rice paddies (Chen et al., 1997; Zheng et al., 2000; Yao et al., 2013). The mean N_2O emission rates were 0.167–0.435 mg N_2O-N m^{-2} d^{-1} during the N fertilized rice cropping period, which was slightly higher than 0.154–0.269 mg N_2O-N m^{-2} d^{-1}

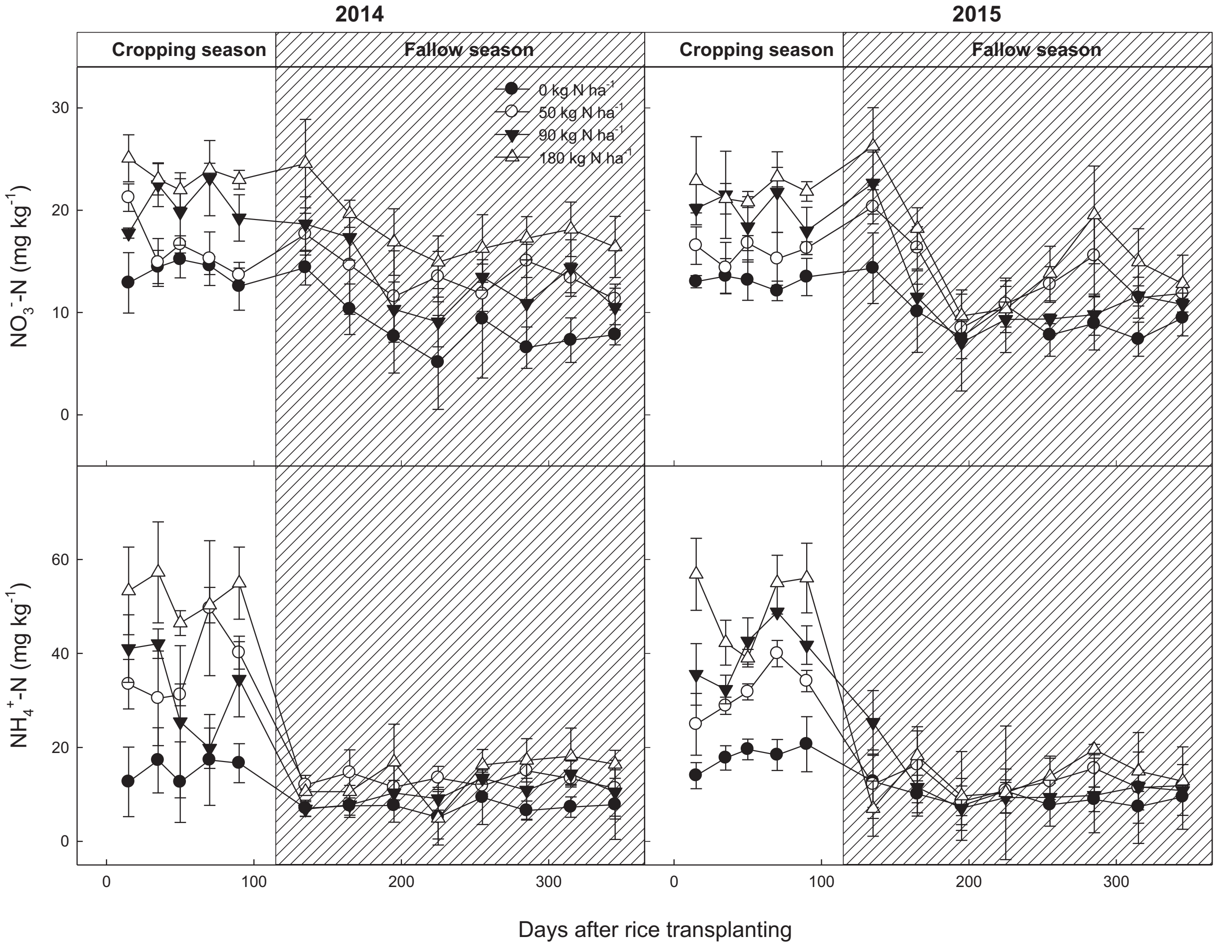

Fig. 3. Changes in inorganic N (NO_3^--N and NH_4^+-N) content in soil with different N fertilizer application levels in the cropping and fallow seasons during the 2-year study.

during the unfertilized fallow season under the same N treatment (Fig. 2). However, irrespective of the N application levels, the seasonal N_2O fluxes were much higher during the long, arid fallow season than during the short waterlogged season (Fig. 4); a similar result was found in a previous study (Haque et al., 2015). For example, in the control plot, the seasonal N_2O fluxes during the fallow season (0.355–0.367 kg N_2O-N ha^{-1}) were approximately 62–74% higher than that during the rice cropping season (0.200–0.208 kg N_2O-N ha^{-1}).

The N_2O EF for rice cropping seasons ranged from 0.0015 to 0.0017 kg N_2O-N kg^{-1} N, which is quite lower than the IPCC default value (0.003 kg N_2O-N kg^{-1} N) (IPCC, 2006). In particular, the N_2O EF was significantly higher on an annual basis, rather than being based

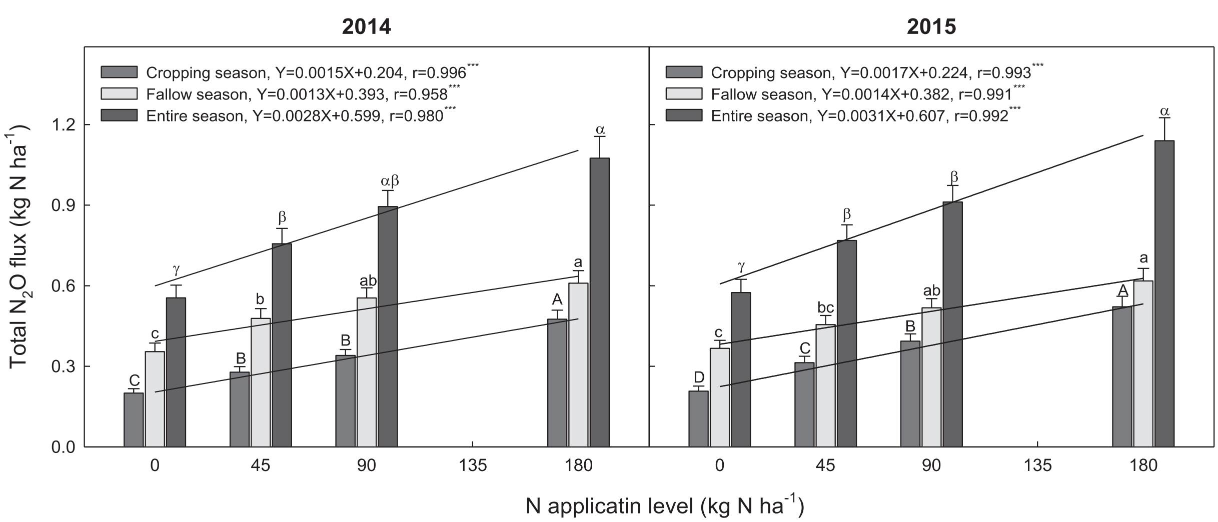

Fig. 4. Total N_2O fluxes and N_2O emission factors under different N application rates during cropping, fallow and the entire season (different letters denote significant differences at the $p < 0.05$ level).

Table 1
Comparison of N_2O emission factors and IPCC default values at a mono-rice field during the cropping, dry fallow and entire season.

Year	Category	Emission factor (kg N_2O N/kg N)	Comparison with IPCC (%)
	IPCC default value	0.003	–
2014	Cropping season	0.0015	46.3
	Fallow season	0.0013	50.0
	Entire season	0.0028	93.3
2015	Cropping season	0.0017	56.7
	Fallow season	0.0014	46.7
	Entire season	0.0031	103.3

Table 2
Correlation between N_2O emissions and soil parameters during a 2-year (cropping and fallow seasons) study of a rice paddy field.

Years	Seasons	Soil parameters	Correlations efficiency (r)
			N_2O emissions
2014	Cropping	Soil temperature	0.339**
		Eh value	−0.216*
		NO_3^--N	0.767***
		NH_4^+-N	0.662**
	Fallow	Soil temperature	0.718***
		Moisture content	0.607***
		NO_3^--N	0.585***
		NH_4^+-N	0.473**
2015	Cropping	Soil temperature	0.334**
		Eh value	−0.211*
		NO_3^--N	0.718***
		NH_4^+-N	0.602**
	Fallow	Soil temperature	0.591***
		Moisture content	0.497***
		NO_3^--N	0.454***
		NH_4^+-N	0.448**

* Significantly correlated at $p < 0.05$.
** Significantly correlated at $p < 0.01$.
*** Significantly correlated at $p < 0.001$.

only on the rice cropping season (Table 1). However, many direct N_2O EFs were developed based only on the cropping season, thus ignoring the N_2O flux during the fallow season (Cai et al., 1997; Kumar et al., 2000; Majumdar et al., 2000; Xia et al., 2016; Zhang et al., 2010). In this study, the fallow season contributed largely to the annual N_2O fluxes, while the contribution during the cropping season was very low. Similar results were observed in Chinese rice paddies, wherein the N_2O EF was statistically estimated using 71 measurements from 17 experimental field data, which was 0.02% of the N applied in continuously flooded paddies during the cropping season (Zou et al., 2007). However, significantly higher N_2O emissions were observed during the dry fallow season than the irrigated rice cropping season in mono-rice paddy fields (Zhou et al., 2014). Therefore, the N_2O EF developed by seasonal N_2O fluxes could be much lower than the annual N_2O EF in temperate mono-rice paddy fields.

As rice paddy soils occupy approximately 46% of the total agricultural land (0.79 million ha) in Korea (Statistics Korea, 2016), it is important to evaluate the N_2O EF for rice paddies to update the information gathered during the Korean GHG inventory. Our newly developed N_2O EF values (0.0028–0.0031 kg N_2O-N kg^{-1}), based on the entire season, were very similar to those of the IPCC default values (0.003 kg N_2O-N kg^{-1} N) of N_2O EF for synthetic N fertilizer application in rice paddy soil (IPCC, 2014). Based on the results of this study, we suggest that while developing the national inventories for GHG emission for rice cropping systems, the N_2O EF should not be confined to the cropping season alone but be based on both the fallow and cropping seasons in the mono-rice paddy field.

5. Conclusion

In a temperate mono-rice paddy, the total N_2O fluxes were significantly increased by N fertilizer application at a rate of 0.0029–0.0045 kg N_2O-N kg N^{-1}, which came from the 36.1–45.8% and 54.2–63.9% seasonal N_2O fluxes during the rice cropping and fallow seasons, respectively. The annual N_2O EF ranged within 0.0028–0.0031 kg N_2O-N kg N^{-1}, similar to the default value (0.003 kg N_2O-N kg N^{-1}) of the IPCC. However, the rice cropping season N_2O EF was only 0.0015–0.0017 kg N_2O-N kg N^{-1}, much lower than the annual N_2O EF. Therefore, we suggest evaluating the N_2O EF based on the entire season and not the cropping season alone.

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